

Effect of magnetic field on the spin resonance in $\text{FeTe}_{0.5}\text{Se}_{0.5}$ as seen via inelastic neutron scattering

Jinsheng Wen,^{1,2} Guangyong Xu,¹ Zhijun Xu,^{1,3} Zhi Wei Lin,¹ Qiang Li,¹ Ying Chen,⁴ Songxue Chi,⁴ Genda Gu,¹ and J. M. Tranquada¹

¹*Condensed Matter Physics and Materials Science Department,
Brookhaven National Laboratory, Upton, New York 11973, USA*

²*Department of Materials Science and Engineering, Stony Brook University, Stony Brook, New York 11794, USA*

³*Physics Department, The City College of New York, New York City, New York 10031, USA*

⁴*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA*

(Dated: March 30, 2010)

Inelastic neutron scattering and susceptibility measurements have been performed on the optimally-doped Fe-based superconductor $\text{FeTe}_{0.5}\text{Se}_{0.5}$, which has a critical temperature, T_c of 14 K. The magnetic scattering at the stripe antiferromagnetic wave-vector $\mathbf{Q} = (0.5, 0.5)$ exhibits a “resonance” at ~ 6 meV, where the scattering intensity increases abruptly when cooled below T_c . In a 7-T magnetic field parallel to the a - b plane, T_c is slightly reduced to ~ 12 K, based on susceptibility measurements. The resonance in the neutron scattering measurements is also affected by the field. The resonance intensity under field cooling starts to rise at a lower temperature ~ 12 K, and the low temperature intensity is also reduced from the zero-field value. Our results provide clear evidence for the intimate relationship between superconductivity and the resonance measured in magnetic excitations of Fe-based superconductors.

PACS numbers: 61.05.fg, 74.70.Dd, 75.25.+z, 75.30.Fv

The recent discovery of Fe-based superconductors^{1–6} has triggered tremendous interest in the field. One of the key questions to be answered is what is the pairing mechanism for the high critical temperature (high- T_c) superconductivity in these materials. It is now widely believed that pairing mediated by magnetic excitations is the most likely candidate for explaining the superconductivity.^{7–12} The “resonance” in magnetic excitations, where the spectral weight at the resonance energy shows a significant increase when the system enters the superconducting phase, has been observed in a number of these Fe-based superconductors, including BaFe_2As_2 (the 1:2:2 system)^{13–17} and the 1:1 system $\text{Fe}_{1+\delta}\text{Te}_{1-x}\text{Se}_x$ ^{18,19}. The resonance is always observed at the energy $\hbar\Omega_0 \sim 5k_B T_c$, and near the antiferromagnetic $(0.5, 0.5)$ point (using notation with two Fe atoms per unit cell) although the propagating vectors for the spin-density-wave (SDW) in the parent compounds are different by 45° in these two systems.^{20–22} These results suggest that the resonance in the magnetic excitations should be similar across different Fe-based superconductor systems, and are closely related to the onset of superconductivity.

In these superconductors, angle resolved photoemission (ARPES) studies^{23–25} have provided evidence for electron and hole pockets that are nearly nested by the stripe antiferromagnetic wave-vector.^{7,26,27} A spin resonance detectable by neutron scattering is predicted to occur at a particular wave-vector only if that wave-vector connects portions of the Fermi surface that have opposite signs of the superconducting gap, so that observations of the resonance may provide important information relevant to the symmetry of the superconducting gap.^{28,29} Since superconductivity, and hence the pairing, is sensitive to magnetic field, one would naturally expect that an external magnetic field can also impact the resonance accordingly, as seen in $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ (Ref. 30) and in $\text{La}_{1.82}\text{Sr}_{0.18}\text{CuO}_4$ (Ref. 31). Indeed, the magnetic field effect on the resonance in Fe-based superconductors has been observed in the 1:2:2

system $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$,³² where the resonance energy and intensity have been partially reduced by an external field.

We have carried out an inelastic neutron scattering study on an optimally-doped 1:1 material—a single crystal of $\text{FeTe}_{0.5}\text{Se}_{0.5}$, with $T_c \approx 14$ K. We find that a resonance with energy $\hbar\Omega_0 \approx 6$ meV $= 5k_B T_c$ appears below T_c , consistent with previous findings.^{18,19} In a 7-T magnetic field parallel to the a - b plane, the superconductivity is partially suppressed, with reduced T_c of 12 K. In the field, the resonance starts to appear at the reduced T_c , with lower intensity than that measured in zero field. This behavior demonstrates that the magnetic excitations have a close association with the superconductivity.

The single-crystal sample was grown by a unidirectional solidification method with nominal composition of $\text{FeTe}_{0.5}\text{Se}_{0.5}$. The bulk susceptibility was characterized using a superconducting quantum interference device (SQUID) magnetometer. In the susceptibility measurements, the sample was oriented so that a - b plane was parallel to the magnetic field. Neutron scattering experiments were carried out on the triple-axis spectrometer BT-7 located at the NIST Center for Neutron Research. A single crystal with mass of 8.9 g was used in the neutron experiment and firmly fixed to an aluminum plate. The lattice constants are $a = b = 3.80(8)$ Å, and $c = 6.14(7)$ Å using the notation where there are two Fe atoms in one unit cell. The data were collected in (HHL) scattering plane, defined by two vectors $[110]$ and $[001]$, and described in reciprocal lattice units (r.l.u.) of $(a^*, b^*, c^*) = (2\pi/a, 2\pi/b, 2\pi/c)$. A vertical magnetic field of 7 T was applied parallel to the a - b plane (along $[1\bar{1}0]$) in the field-cooling (FC) measurements.

Energy scans have been performed at $\mathbf{Q} = (0.5, 0.5, 0)$, as shown in Fig. 1(a). There is a large background at low energies coming from the superconducting magnet in which the sample resides, and this obscures the magnetic response in the raw data. However, if we compare the scans taken at 4 K and

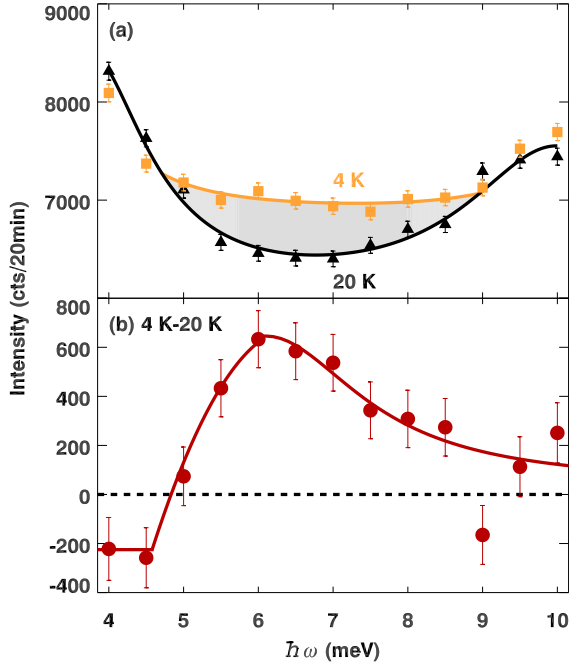


FIG. 1: (Color online) (a) Constant \mathbf{Q} scans at $(0.5, 0.5, 0)$ for temperatures below ($T = 4$ K) and above ($T = 20$ K) T_c . Shading indicates the difference between scans. (b) Data obtained by subtracting 20 K data from 4 K data. Error bars represent square root of total counts. Lines through data are guides for the eye.

20 K, a significant amount of spectral weight shows up between 5 meV and 9 meV for the spectrum measured at low temperature (as indicated by the shading). If we subtract the 20 K data from the 4 K data as in Fig. 1(b), one can see a broad peak at ~ 6 meV. This is consistent with that observed in 40% and 50% Se doped samples, in which resonance energies of 6.5 meV and 7 meV, respectively, were reported.^{18,19} Although a spin gap is not directly observed in the raw data, we do see from the background subtracted data in Fig. 1(b) that the difference of the intensity ($I_{4K} - I_{20K}$) becomes negative below 5 meV, which suggests that a gap opens below this energy at 4 K, consistent with the gap value obtained by Qiu *et al.*¹⁸

To test the impact of a magnetic field, a 7-T field was applied at 20 K, and the sample was cooled in the field. In Fig. 2, we show background (20 K data, zero field) subtracted scans performed at different temperatures. At $T = 12$ K, the difference between data taken with and without the field is very clear. With further cooling, the difference is still observable but becomes less pronounced. At $T = 4$ K, the peak intensity for the 7-T scan is about 10% to 20% smaller than that of the zero-field data, while the 7-T spectrum seems to have more intensity filled in below the gap (~ 5 meV).

We also performed some constant-energy (7-meV) scans along $(h, h, 0)$ through $h = 0.5$. With a counting rate of 5 min/point, the change in signal at $h \approx 0.5$ between 4 K and 20 K was consistent with the constant- \mathbf{Q} scans; however, the signal-to-background level at this counting rate was not sufficient to provide a useful measure of the peak shape, nor to

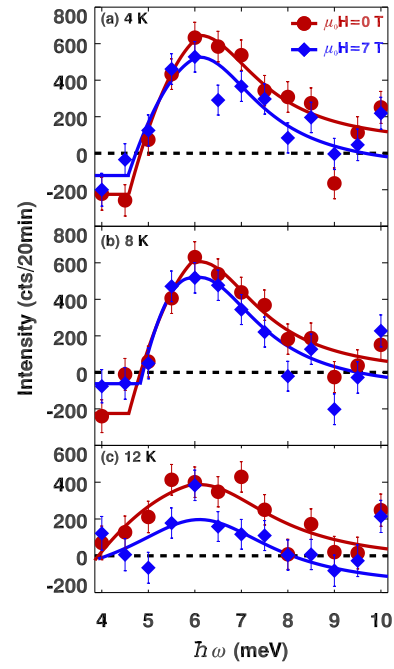


FIG. 2: (Color online) Constant \mathbf{Q} scans at $(0.5, 0.5, 0)$, after subtraction of the zero-field scan at 20 K. (a) $T = 4$ K, (b) 8 K, (c) 12 K, for $\mu_0 H = 0$ T (circles), and 7 T (diamonds). Error bars represent square root of total counts. Lines through data are guides for the eye.

resolve changes due to field. Given finite beam time, it was not possible to measure both constant- \mathbf{Q} and constant-energy scans with adequate statistics, so we decided to abandon the latter.

There is a sum rule for scattering from spin-spin correlations, and hence one might expect that the reduction of the resonance intensity by the field should result in an increase of spectral weight below the gap, as commonly seen in cuprates,^{33–36} as well as in $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$ (Ref. 32). As discussed above, it is consistent with our results in principle, but the large background makes it impossible to follow the behavior to lower energies. In cuprates, Demler *et al.* analyzed a model of coexisting but competing phases of superconductivity and SDW order,³⁷ and successfully predicted the field-induced static magnetic order observed experimentally.^{38–40} We have searched for SDW order around $(0.5, 0.5, 0)$, but no evidence of such field-induced order was found.

We have measured the bulk susceptibility in 0-T and 7-T field as well, and the results are shown in Fig. 3(a). In zero field, the system enters a superconducting state at 14 K, and becomes fully diamagnetic below 12 K. In the 7-T field, superconductivity is partially suppressed, and T_c has been reduced to 12 K. As a result of the suppressed superconductivity, the resonance intensity has also been reduced as shown in Fig. 2.

Fig. 3(b) gives another perspective of the impact of the field on the resonance. There we plot the intensity, integrated from 6 meV to 7 meV, as a function temperature obtained for the measurements with and without the field. The intensity $I(T)$ was fit with the mean-field theory⁴¹ using T_c s determined by the onset of the diamagnetism in Fig. 3(a), with

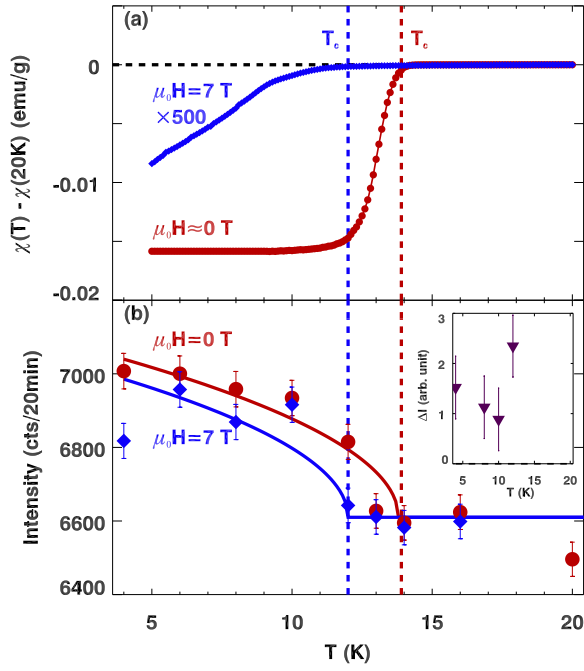


FIG. 3: (Color online) (a) Susceptibility measured with $\mu_0 H = 0.0005$ T (circles) and 7 T (diamonds), with field parallel to the a - b plane. Dashed lines indicate the T_c s. (b) Resonance intensity at (0.5, 0.5, 0) integrated from 6 meV to 7 meV. The solid lines are fits using mean-field theory (described in the text), with T_c s obtained from (a). Inset shows the difference of the resonance intensities for 0 T and 7 T, integrated from 5 meV to 8 meV. Error bars represent square root of total counts.

$I(T) = I(0)(1 - T/T_c)^{1/2} + A$, where $I(0)$, and A are constants. This formula results in the solid lines, which fit the data reasonably well. In both 0 T and 7 T, the resonance intensity starts to appear below respective T_c , and increases with cooling. At low temperatures, the intensity at 7 T is lower than the zero-field value. To confirm that the intensity is reduced at 7 T, we plot in the inset of Fig. 3(b) the difference between intensity at 0 T and 7 T, ΔI , integrated from 5 meV to 8 meV; one can see that the intensity difference is well above zero.

With Fig. 3, one can better understand the results in Fig. 2, especially the most pronounced field effect at 12 K. In zero field, the sample is in superconducting state at 12 K, where the resonance has finite intensity; in the 7-T field, the system is driven to normal state at this temperature, and the resonance intensity is approaching background level.

From the data, it is clear that the magnetic field depresses the superconductivity, and also reduces the onset temperature and intensity of the resonance. In principle, if the resonance is directly associated with the superconducting volume of the

sample, the intensity ratio I_{7T}/I_{0T} should be roughly proportional to $1 - H/H_{c2}$, where H is the applied field, and H_{c2} is the upper critical field.³⁰ Our results showing a change $\sim 10\%$ in the resonance intensity, suggesting that H_{c2} is ~ 70 T, which is comparable to the range estimated in other studies.^{42,43} Although no significant change in the resonance with field was identified for the 40% Se sample in Ref. 18, we believe that our results are consistent with that study within the error bars. The fact that the field also suppresses the resonance intensity in $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$ ³² suggests that this should be common in Fe-based superconductors.

There are of course, still issues not fully resolved based on our results. For example, the quality of our data does not allow us to accurately determine the resonance energy. It is therefore hard to find out whether the resonance energy can be affected by the external magnetic field or not, although it has been shown that the former is the case in $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$.³² We have measured the susceptibility with field perpendicular to a - b plane, and compared it with the data in this work.⁴⁴ It is shown that there is only anisotropy in the superconducting state. It will be interesting to see how the resonance responds to a c -axis magnetic field. Another interesting issue is to search for the Zeeman splitting of the resonance mode under an external field, which is a good test of whether this is a singlet-triplet excitation. Zhao *et al.*³² tried to tackle this problem using a 14.5-T field, but the results are inconclusive—the resonance in $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$ broadens in the field, but no clear split was observed, probably due to the finite resonance width and coarse energy resolution. Qiu *et al.*¹⁸ applied a 7-T magnetic field on $\text{FeTe}_{0.6}\text{Se}_{0.4}$, but no splitting is visible from their results; in a more recent experiment, with a larger field (14 T) and improved background, they were able to resolve the Zeeman splitting, directly establishing its triplet character⁴⁵.

In summary, we observed a resonance at $\hbar\Omega_0 \approx 6$ meV in $\text{FeTe}_{0.5}\text{Se}_{0.5}$ ($T_c = 14$ K). The temperature dependence of the intensity is consistent with the scaling $1 - (T/T_c)^{1/2}$. A 7 T magnetic field partially suppresses superconductivity, and lowers T_c to about 12 K, determined from the bulk susceptibility. In the field, the resonance starts to appear at the lowered T_c , 12 K, with intensity reduced. These results are consistent with the picture that the resonance is related to quasiparticle scattering in the superconducting phase, and is reduced when superconductivity becomes weaker, either by heating or applying an external magnetic field.

The work at Brookhaven National Laboratory was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, under Contract No. DE-AC02-98CH10886.

¹ Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. **130**, 3296 (2008).

² H. Takahashi, K. Igawa, K. Arii, Y. Kamihara, M. Hirano, and H. Hosono, Nature **453**, 376 (2008).

³ X. H. Chen, T. Wu, G. Wu, R. H. Liu, H. Chen, and D. F. Fang, Nature **453**, 761 (2008).

⁴ Z.-A. Ren, W. Lu, J. Yang, W. Yi, X.-L. Shen, Z.-C. Li, G.-C. Che, X.-L. Dong, L.-L. Sun, F. Zhou, et al., Chin. Phys. Lett. **25**,

- 2215 (2008).
- ⁵ F.-C. Hsu, J.-Y. Luo, K.-W. Yeh, T.-K. Chen, T.-W. Huang, P. M. Wu, Y.-C. Lee, Y.-L. Huang, Y.-Y. Chu, D.-C. Yan, et al., *Proc. Natl. Acad. Sci. U.S.A.* **105**, 14262 (2008).
 - ⁶ K.-W. Yeh, T.-W. Huang, Y.-L. Huang, T.-K. Chen, F.-C. Hsu, P. M. Wu, Y.-C. Lee, Y.-Y. Chu, C.-L. Chen, J.-Y. Luo, et al., *Europhys. Lett.* **84**, 37002 (2008).
 - ⁷ I. I. Mazin, D. J. Singh, M. D. Johannes, and M. H. Du, *Phys. Rev. Lett.* **101**, 057003 (2008).
 - ⁸ K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kontani, and H. Aoki, *Phys. Rev. Lett.* **101**, 087004 (2008).
 - ⁹ F. Ma and Z.-Y. Lu, *Phys. Rev. B* **78**, 033111 (2008).
 - ¹⁰ J. Dong, H. J. Zhang, G. Xu, Z. Li, G. Li, W. Z. Hu, D. Wu, G. F. Chen, X. Dai, J. L. Luo, et al., *Europhys. Lett.* **83**, 27006 (2008).
 - ¹¹ V. Cvetkovic and Z. Tesanovic, *Europhys. Lett.* **85**, 37002 (2009).
 - ¹² S. Graser, T. A. Maier, P. J. Hirschfeld, and D. J. Scalapino, *New J. Phys.* **11**, 025016 (2009).
 - ¹³ A. D. Christianson, E. A. Goremychkin, R. Osborn, S. Rosenkranz, M. D. Lumsden, C. D. Malliakas, I. S. Todorov, H. Claus, D. Y. Chung, M. G. Kanatzidis, et al., *Nature* **456**, 930 (2008).
 - ¹⁴ M. D. Lumsden, A. D. Christianson, D. Parshall, M. B. Stone, S. E. Nagler, G. J. MacDougall, H. A. Mook, K. Lokshin, T. Egami, D. L. Abernathy, et al., *Phys. Rev. Lett.* **102**, 107005 (2009).
 - ¹⁵ S. Chi, A. Schneidewind, J. Zhao, L. W. Harriger, L. Li, Y. Luo, G. Cao, Z. Xu, M. Loewenhaupt, J. Hu, et al., *Phys. Rev. Lett.* **102**, 107006 (2009).
 - ¹⁶ S. Li, Y. Chen, S. Chang, J. W. Lynn, L. Li, Y. Luo, G. Cao, Z. Xu, and P. Dai, *Phys. Rev. B* **79**, 174527 (2009).
 - ¹⁷ D. S. Inosov, J. T. Park, P. Bourges, D. L. Sun, Y. Sidis, A. Schneidewind, K. Hradil, D. Haug, C. T. Lin, B. Keimer, et al., *Nature Phys.* **6**, 178 (2010).
 - ¹⁸ Y. Qiu, W. Bao, Y. Zhao, C. Broholm, V. Stanev, Z. Tesanovic, Y. C. Gasparovic, S. Chang, J. Hu, B. Qian, et al., *Phys. Rev. Lett.* **103**, 067008 (2009).
 - ¹⁹ H. A. Mook, M. D. Lumsden, A. D. Christianson, B. C. Sales, R. Jin, M. A. McGuire, A. Sefat, D. Mandrus, S. E. Nagler, T. Egami, et al., arXiv:0904.2178.
 - ²⁰ W. Bao, Y. Qiu, Q. Huang, M. A. Green, P. Zajdel, M. R. Fitzsimmons, M. Zhernenkov, M. Fang, B. Qian, E. K. Vehstedt, et al., *Phys. Rev. Lett.* **102**, 247001 (2009).
 - ²¹ S. Li, C. de la Cruz, Q. Huang, Y. Chen, J. W. Lynn, J. Hu, Y.-L. Huang, F.-C. Hsu, K.-W. Yeh, M. Kuen Wu, et al., *Phys. Rev. B* **79**, 054503 (2009).
 - ²² Jinsheng Wen, Guangyong Xu, Zhijun Xu, Zhi Wei Lin, Qiang Li, W. Ratcliff, G. Gu, and J. M. Tranquada, *Phys. Rev. B* **80**, 104506 (2009).
 - ²³ C. Liu, G. D. Samolyuk, Y. Lee, N. Ni, T. Kondo, A. F. Santander-Syro, S. L. Bud'ko, J. L. McChesney, E. Rotenberg, T. Valla, et al., *Phys. Rev. Lett.* **101**, 177005 (2008).
 - ²⁴ V. B. Zabolotnyy, D. S. Inosov, D. V. Evtushinsky, A. Koitzsch, A. A. Kordyuk, G. L. Sun, J. T. Park, D. Haug, V. Hinkov, A. V. Boris, et al., *Nature* **457**, 569 (2009).
 - ²⁵ K. Terashima, Y. Sekiba, J. H. Bowen, K. Nakayama, T. Kawahara, T. Sato, P. Richard, Y.-M. Xu, L. J. Li, G. H. Cao, et al., *Proc. Natl. Acad. Sci. U.S.A.* **106**, 7330 (2009).
 - ²⁶ K. Seo, B. A. Bernevig, and J. Hu, *Phys. Rev. Lett.* **101**, 206404 (2008).
 - ²⁷ I. I. Mazin and J. Schmalian, *Physica C* **469**, 614 (2009).
 - ²⁸ T. A. Maier and D. J. Scalapino, *Phys. Rev. B* **78**, 020514(R) (2008).
 - ²⁹ T. A. Maier, S. Graser, D. J. Scalapino, and P. Hirschfeld, *Phys. Rev. B* **79**, 134520 (2009).
 - ³⁰ P. Dai, H. A. Mook, G. Aeppli, S. M. Hayden, F. Doğan, J. Yu, Y. Yanagida, H. Takashima, Y. Inaguma, M. Itoh, et al., *Nature* **406**, 965 (2000).
 - ³¹ J. M. Tranquada, C. H. Lee, K. Yamada, Y. S. Lee, L. P. Regnault, and H. M. Rønnow, *Phys. Rev. B* **69**, 174507 (2004).
 - ³² J. Zhao, L.-P. Regnault, C. Zhang, M. Wang, Z. Li, F. Zhou, Z. Zhao, and P. Dai, arXiv:0908.0954.
 - ³³ B. Lake, G. Aeppli, K. N. Clausen, D. F. McMorrow, K. Lefmann, N. E. Hussey, N. Mangkorntong, M. Nohara, H. Takagi, T. E. Mason, et al., *Science* **291**, 1759 (2001).
 - ³⁴ M. Kofu, S. H. Lee, M. Fujita, H. J. Kang, H. Eisaki, and K. Yamada, *Phys. Rev. Lett.* **102**, 047001 (2009).
 - ³⁵ Jinsheng Wen, Zhijun Xu, Guangyong Xu, J. M. Tranquada, Genda Gu, S. Chang, and H. J. Kang, *Phys. Rev. B* **78**, 212506 (2008).
 - ³⁶ S. D. Wilson, S. Li, J. Zhao, G. Mu, H.-H. Wen, J. W. Lynn, P. G. Freeman, L.-P. Regnault, K. Habicht, and P. Dai, *Proc. Natl. Acad. Sci. U.S.A.* **104**, 15259 (2007).
 - ³⁷ E. Demler, S. Sachdev, and Y. Zhang, *Phys. Rev. Lett.* **87**, 067202 (2001).
 - ³⁸ B. Lake, H. M. Rønnow, N. B. Christensen, G. Aeppli, K. Lefmann, D. F. McMorrow, P. Vorderwisch, P. Smeibidl, N. Mangkorntong, T. Sasagawa, et al., *Nature* **415**, 299 (2002).
 - ³⁹ D. Haug, V. Hinkov, A. Suchaneck, D. S. Inosov, N. B. Christensen, C. Niedermayer, P. Bourges, Y. Sidis, J. T. Park, A. Ivanov, et al., *Phys. Rev. Lett.* **103**, 017001 (2009).
 - ⁴⁰ B. Lake, K. Lefmann, N. B. Christensen, G. Aeppli, D. F. McMorrow, H. M. Rønnow, P. Vorderwisch, P. Smeibidl, N. Mangkorntong, T. Sasagawa, et al., *Nature Mater.* **4**, 658 (2005).
 - ⁴¹ N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Brooks Cole, 1976), Chap. 34, P. 744.
 - ⁴² W. Si, Z.-W. Lin, Q. Jie, W.-G. Yin, J. Zhou, G. Gu, P. D. Johnson, and Q. Li, *Appl. Phys. Lett.* **95**, 052504 (2009).
 - ⁴³ T. Kida, T. Matsunaga, M. Hagiwara, Y. Mizuguchi, Y. Takano, and K. Kindo, *J. Phys. Soc. Jpn* **78**, 113701 (2009).
 - ⁴⁴ Jinsheng Wen, Guangyong Xu, Zhijun Xu, G. Gu, and J. M. Tranquada, unpublished.
 - ⁴⁵ W. Bao, A. T. Savici, G. E. Granroth, C. Broholm, K. Habicht, Y. Qiu, J. Hu, T. Liu, and Z. Q. Mao, arXiv:1002.1617.